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EP 0 533 493 B1

Description

BACKGROUND OF THE INVENTION

5 The present invention relates to laminated films each constituted by at least two layers. More particularly, the present invention relates to laminated films having flexibility, mechanical strengths, transparency, surface wettability, low-temperature sealability, etc., which are each a soft film comparable to a soft vinyl chloride resin film and a vinylon film.

10 In recent years, soft vinyl chloride resin films containing a plasticizer have been widely used as a soft resin film. Soft vinyl chloride resins, however, may give rise to social problems such as (1) toxicity caused by bleed-out of plasticizer of monomer used therein, (2) transition and (3) acid rain derived from hydrogen chloride generated by burning thereof.

Meanwhile, as a soft resin film similar to the soft vinyl chloride resin films, there are resin films using ethylene as a main component, such as ethylene/vinyl acetate copolymer film, low-density polyethylene film, ionomer film and the like. These ethylene-based soft resin films, however, are inferior to soft vinyl chloride resin films in transparency, haze, 15 glass, etc, and moreover have poor impact strength, heat resistance, stiffness.

A vinylon film widely used for fiber packaging has excellent feeling of flexibility but, unlike polyolefin films, is extremely costly.

Meanwhile, in crystalline polypropylene films, it is possible to impart flexibility while maintaining their transparency and haze, by using a propylene random copolymer of reduced melting point obtained by randomly copolymerizing propylene and ethylene and/or an α -olefin. With any existing technique, however, it is difficult to allow a crystalline polypropylene resin to have flexibility comparable to that of soft vinyl chloride resin. 20

DE-A-2815855 discloses a flexible packaging film comprising a biaxially-oriented layer of isotactic polypropylene and a non-oriented layer of a mixture of isotactic and atactic polypropylene, the two layers being bonded together using an adhesive.

25 The present invention is intended to provide a resin film which has good transparency, haze, gloss, etc., which has excellent flexibility and mechanical strength, and which has good impact resistance, surface wettability, low-temperature sealability, etc.

The present invention is also intended to provide a resin film suitable for use, because of the above properties, in various applications, for example, a packaging film on which multi-colored decorative printing has been made (e.g. food-packaging film, fiber-packaging film), a transfusion bag and intravenous bag film used in medical care, a metal-protecting film, an adhesive film, and a building material film for use in civil engineering and construction. 30

SUMMARY OF THE INVENTION

35 According to the present invention, a laminated film obtainable by melt coextrusion of at least 2 layers comprises:

a layer (A) containing a fibre-free composition comprising (i) 30 to 100% by weight of an amorphous polyolefin derived from, by weight of the polyolefin, at least 50% of propylene and/or butene-1, and (ii) 70 to 0% by weight of a crystalline polypropylene; and

40 an outermost layer (B) comprising (i) a crystalline polypropylene, (ii) an ethylene (co)polymer or (iii) a blend of a crystalline polypropylene and a copolymer of ethylene and a vinyl monomer having a polar group, or a polymer that is obtainable by grafting a vinyl monomer having a polar group to an ethylene (co)polymer.

In the following, when layer B comprises a crystalline polypropylene (i) it is referred to as B₁; when layer B comprises an ethylene (co)polymer (ii) it is referred to as B₂; and when layer B comprises a blend of a crystalline polypropylene and a copolymer of ethylene and a vinyl monomer having a polar group, or a polymer that is obtainable by grafting a vinyl monomer having a polar group to an ethylene (co)polymer (iii) it is referred to as B₃. 45

By subjecting to composite lamination (e.g. co-extrusion or lamination) a layer comprising an amorphous polyolefin containing propylene and a crystalline polypropylene and a layer comprising a crystalline polypropylene, there can be obtained a laminated film having excellent transparency, haze and gloss, retaining the mechanical strengths inherently possessed by polypropylene, and having high flexibility. 50

By subjecting to the same composite lamination as above a layer comprising an amorphous polyolefin containing propylene and/or butene-1 and a crystalline polypropylene and a layer comprising an ethylene (co)polymer resin, there can be obtained a laminated film having the above properties and excellent low-temperature sealability.

55 By subjecting to the same treatment as above a layer comprising an amorphous polyolefin containing propylene and/or butene-1 and a crystalline polypropylene and a layer comprising a crystalline polypropylene and an ethylene (co)polymer resin having a polar group, there can be obtained a laminated film having the above properties and excellent surface wettability.

DETAILED DESCRIPTION OF THE INVENTION

The present inventors made study in order to achieve the above objects. As a result, the present inventors found that the above objects can be met by laminated films each constituted by a layer comprising an amorphous polyolefin of particular composition or said amorphous polyolefin and a crystalline polypropylene at a desired ratio and a layer which is an appropriate combination of a crystalline polypropylene, an ethylene (co)polymer resin and an ethylene (co)polymer resin having a polar group. The finding has led to the completion of the present invention.

The laminated films of the present invention are described in detail below.

In the laminated films of the present invention, the amorphous polyolefin used in the layer A (hereinafter referred to as amorphous polyolefin) can be any amorphous olefin polymer containing propylene and/or butene-1 in an amount of 50% by weight or more. There can be used, for example, an amorphous polypropylene, a polybutene-1, or a copolymer of propylene and/or butene-1 and other α -olefin. When the propylene and/or butene-1 content in the amorphous polyolefin is less than 50% by weight, the amorphous polyolefin has low compatibility with a crystalline polypropylene; hence, such a propylene and/or butene-1 content is not desirable.

The amorphous polyolefin typically has a boiling-n-heptane insoluble content (i.e. a boiling-n-heptane when subjected to Soxhlet extraction) of 70% by weight or less, preferably 60% by weight or less. When the boiling-n-heptane insoluble is more than 70% by weight, the proportion of the amorphous portion in the amorphous polyolefin is small, making it impossible to obtain a resin film having desired sufficient flexibility. The amorphous polyolefin in the layer A further typically has a number-average molecular weight of preferably 1,000-200,000, more preferably 1,500-100,000. When the number-average molecular weight is more than 200,000, film molding is difficult. When the number-average molecular weight is less than 1,000, the resulting film has low mechanical strengths. In the present invention, the amorphous polyolefin can be used singly or in combination of two or more kinds.

As the amorphous polyolefin, there may be used an atactic polypropylene which is produced as a by-product in producing a crystalline polypropylene. Alternatively, the amorphous polyolefin may be produced as such from a raw material. The copolymer of propylene and/or butene-1 and other α -olefin can be produced as an intended product from raw materials so as to contain propylene and/or butene-1 in a desired proportion.

When the amorphous polyolefin is produced as an intended product, it can be obtained by, for example, polymerizing raw material monomers using (a) a titanium catalyst supported on magnesium chloride and (b) triethylaluminum, in the presence or absence of hydrogen. Use of an amorphous polyolefin produced as an intended product is preferable in view of its stable supply and stable quality. As the amorphous polyolefin, there may be used a commercial product if there is an appropriate commercial product.

Specific examples of the amorphous polyolefin used in the layer A of the present invention include those containing propylene as a main component in a given proportion and having desired properties, such as polypropylene, propylene/ethylene copolymer, propylene/butene-1 copolymer, propylene/hexene-1 copolymer, propylene/butene-1/ethylene terpolymer, propylene/hexene-1/octene-1 terpolymer, propylene/hexene-1/4-methylpentene-1 terpolymer and the like.

Specific examples of the amorphous polyolefin also include those containing butene-1 in a given proportion and having desired properties, such as polybutene-1, butene-1/ethylene copolymer, butene-1/propylene copolymer, butene-1/propylene/ethylene terpolymer, butene-1/hexene-1/octene-1 terpolymer, butene-1/hexene-1/4-methylpentene-1 terpolymer and the like.

When the amorphous polyolefin is a propylene/ethylene copolymer, the copolymer desirably has an ethylene content of up to 30% by weight, preferably 1-20% by weight. When the ethylene content is larger than 30% by weight, the resulting film is too soft.

When the amorphous polyolefin in the layer A of the present invention is a propylene/butene-1 copolymer, the propylene/butene-1 copolymer can be of three types, (1) a copolymer containing propylene as a main component, (2) a copolymer containing butene-1 as a main component, and (3) a copolymer containing both propylene and butene-1 as its main components.

Each of these copolymers desirably has its main component(s) of 50% by weight or more, the other component(s) of less than 50%, preferably 1-45% by weight, more preferably 5-44% by weight.

The propylene/butene-1 copolymer has high tensile elongation and cohesiveness and can be suitably used as the amorphous polyolefin of the layer A. An example of the commercial products of the propylene/butene-1 copolymer usable in the layer A is REXTAC of Rexene Co. of U.S.A.

The crystalline polypropylene used in the layer A of the present invention refers to an isotactic polypropylene insoluble in boiling n-heptane and includes commercially available polypropylenes used for extrusion, injection molding, blow molding, etc. It may be a propylene homopolymer, or a copolymer between an isotactic polypropylene of stereoregularity and other α -olefin.

As the crystalline polypropylene, there may be used a commercial product or a product produced as such. The production of crystalline polypropylene is not critical and can be conducted using a process appropriately selected from conventional processes used for production of crystalline polypropylene.

The α -olefin used in copolymerization with a crystalline polypropylene is preferably an α -olefin of 2-8 carbon atoms,

such as ethylene, butene-1, pentene-1, hexene-1, heptene-1, octene-1 or the like. Of these, ethylene or butene-1 is particularly preferable.

In the present invention, as the crystalline polypropylene, there are preferably mentioned a propylene homopolymer; a propylene/ethylene random or block copolymer having an ethylene content of 30% by weight or less, preferably 1-25% by weight; a propylene/butene-1 random or block copolymer having a butene-1 content of 20% by weight or less. Of these, a copolymer between ethylene or butene-1 and propylene is particularly preferable in view of the applications (film, sheet, etc.) of the present resin composition. The crystalline polypropylene of the layer A can be used singly or in combination of two or more kinds.

In the present invention, the amorphous polyolefin and/or crystalline polypropylene of the layer A may be used in modified forms (form). That is, the amorphous polyolefin or crystalline polypropylene may be used after being modified with an unsaturated carboxylic acid (e.g. acrylic acid, methacrylic acid, ethacrylic acid, maleic acid, fumaric acid, itaconic acid) and/or its derivative (e.g. ester, acid anhydride, metal salt) or with other unsaturated compound (e.g. amide, amino compound, glycidyl methacrylate, hydroxymethacrylate). Of the modification products, preferable is a product modified with maleic anhydride or itaconic anhydride, and more preferable is a product modified with maleic anhydride.

In the present invention, the production of the resin composition of the layer A containing the amorphous polyolefin and the crystalline polypropylene is not critical and can be carried out by a method ordinarily used in the production of conventional polypropylene compositions, wherein melt kneading is conducted with heating, using, for example, a kneader (e.g. kneader, Banbury mixer, rolls) or a single-screw or twin-screw extruder.

In the present invention, the resin composition constituting the layer A may contain, as necessary, various additives and fillers, such as heat stabilizer, antioxidant, light stabilizer, antistatic agent, lubricant, nucleating agent, flame retardant, pigment or dye, calcium carbonate, calcium sulfate, barium sulfate, magnesium hydroxide, mica, talc, clay and the like. The resin composition may further contain, as necessary, other thermoplastic resins, thermoplastic elastomers, rubbers, etc. It is possible that these resins, elastomers, rubbers, etc. be compounded so as to form a crosslinked structure. When flame retardancy is imparted to the resin composition, magnesium hydroxide or magnesium carbonate is added in an amount of 20-60 parts by weight of laminated film.

The resin composition of the layer A of the present invention comprises 30-100% by weight amorphous polyolefin and 70-0% by weight crystalline polypropylene.

The crystalline polypropylene which can be used in the layer B₁ and/or B₃ of the present invention may be the same crystalline polypropylene as used in the layer A. It is particularly preferably a propylene random copolymer having a meltingpoint of 150° C or less so that the resulting laminated film can have a feature of soft film.

As the ethylene (co)polymer resin which can be used in the layer B₂, there are mentioned a low-density polyethylene, a linear low-density polyethylene, a very-low-density polyethylene, a medium-density polyethylene, a high-density polyethylene and a copolymer containing ethylene as a main component. The copolymer includes copolymers and multi-component polymers between ethylene and at least one comonomer selected from olefins (e.g. propylene, butene, pentene, hexene, heptene, octene), vinyl esters (e.g. vinyl acetate, vinyl propionate), unsaturated carboxylic acid esters (e.g. methyl acrylate, ethyl acrylate, methyl methacrylate, ethyl methacrylate) and salts of metal unsaturated carboxylates thereof. The ethylene (co)polymer resin may be a combination of two or more polymers or copolymers. It may also be a modification product as in the case of the amorphous polyolefin and/or the crystalline polypropylene of the layer A.

The ethylene (co)polymer resin having a polar group which can be used in the layer B₃ may be a copolymer of ethylene and a vinyl monomer having a polar group, or a graft polymer obtained by grafting a vinyl monomer having a polar group, to an ethylene (co)polymer resin.

As the former, there can be used copolymers or multi-component polymers between ethylene and unsaturated acid(s), unsaturated ester(s) or salts of metal unsaturated carboxylates thereof, such as ethylene/vinyl acetate copolymer, ethylene/acrylic acid copolymer, ethylene/ethyl acrylate copolymer, ethylene/methyl methacrylate copolymer, ethylene/glycidyl methacrylate and other ionomers. It is possible to use two or more copolymers or multi-component polymers in combination.

As the latter, there can be used modification products obtained by grafting (modification) with an unsaturated carboxylic acid (e.g. acrylic acid, methacrylic acid, ethacrylic acid, maleic acid, fumaric acid, itaconic acid) and/or a derivative thereof (e.g. ester, acid anhydride, metal salt). Of these modification products, there are preferably used those modified with maleic anhydride or itaconic anhydride, more preferably those modified with maleic anhydride.

The hardness of the ethylene (co)polymer resin used in layer B₂ and/or B₃ is not critical but is preferably 50 or less in terms of Shore D hardness because the layers are used as part of a soft film. As the ethylene (co)polymer resin having a Shore D hardness of 50 or less, there are mentioned a low-density polyethylene, a linear low-density polyethylene, a very-low-density polyethylene, an ethylene/vinyl acetate copolymer, an ethylene/ethyl acrylate copolymer, ionomers, etc.

The ratio of the crystalline polypropylene and the ethylene (co)polymer resin having a polar group, in layer B₃ is not critical but is preferably 98:2 to 50:50, more preferably 95:5 to 60:40 in view of the compatibility of the two components and the transparency of the layer.

The concentration of the monomer having a polar group in the ethylene (co)polymer resin or the concentration of the graft component (e.g. acid) in the ethylene (co)polymer resin is preferably 0.01-30% by weight, more preferably 0.05-20% by weight based on the total resin of the B₃ layer in order for the surface of the resulting film to exhibit desired wettability and adhesivity.

5 When the laminated films of the present invention consist of a number of layers A and/or layers B they are ordinarily constituted in such a manner that the layers A and the layers B are laminated alternately in the above-mentioned combination. The number of the layers A and the layers B used has no restriction, but at least one layer A and at least one layer B are laminated so that at least one of the two outermost layers is a layer B. The laminated films can be constituted in various combinations such as B₁/A, B₂/A/B₂, B₃/A/B₃/A/B₃ and the like. Three-layered laminated films such as
10 B₁/A/B₁ and the like are generally used. It is possible to use a combination of B₁/A/X by laminating X (a resin other than A and B₁) onto the layer A of B₁/A.

In the present invention, the layer A contributes in imparting flexibility to the resulting film. Containing a crystalline polypropylene, the layer A can also endow the resulting film with heat resistance, firmness and elasticity. Meanwhile, the amorphous polyolefin also contained in the layer A has high surface adhesivity; therefore, when an amorphous poly-
15 olefin of low molecular weight is used, the resulting film has striking surface adhesivity.

The B₂ layer contributes in imparting not only the above-mentioned properties but also low-temperature sealability. Therefore, the resulting laminated film has excellent low-temperature sealability. With the layer B₃, the polar group is exposed on the surface; therefore, using the layer B₃, there can be obtained a laminated film making easy adhesive coating and multi-color or complex printing.

20 In the laminated films of the present invention, it is possible to insert, between the layer A and the layer B other thermoplastic resin, for example, a nylon, a poly(vinyl alcohol), a saponified EVA, polyester, etc., in order to impart gas-barrier property.

In the present invention, the thickness of each layer constituting the laminated films is not critical and can be determined as desired. However, each layer is ordinarily produced so as to have a thickness in the range of about 2-1,000
25 mm. The thickness ratio of the layer A and the layer B is not critical, either. However, the total thickness of the layer(s) A is determined so as to be preferably 20-99%, more preferably 30-95% of the total thickness of laminated film in order for the layer A to contribute to film flexibility and for the layer B to serve to suppress surface adhesivity.

The method for producing the laminated films of the present invention has no particular restriction. There can be used, for example, a co-extrusion lamination method, a lamination method, or a dry lamination method. Of these, there
30 is preferred the co-extrusion lamination method wherein melt adhesion is conducted. The co-extrusion lamination is specifically conducted, for example, by carrying out melt extrusion using extruders (the number of the extruders is the same as that of the layers in the laminated film produced), laminating the extrudates in a molten state by a known method such as T-die method, inflation method or the like, then cooling the resulting laminate with cooling rolls, air or water to obtain a laminated film.

35 The laminated films of the present invention can be subjected to a surface treatment in order to improve the printability, laminatability and adhesive coatability. The method for the surface treatment includes a corona discharge treatment, a plasma treatment, a flame treatment, an acid treatment, etc. Any of these methods can be used in the present invention. The plasma treatment, the flame treatment, or the corona treatment is preferable because it enables continuous treatment and can be easily carried out before the winding step. Of them, the corona treatment is most preferable
40 in view of the convenience. The laminated films of the present invention may be stretched monoaxially or biaxially.

The laminated films of the present invention are subjected, after the above-mentioned lamination, cooling and solidification, to the above treatment as necessary, wound, and subjected to secondary steps (e.g. printing, lamination, adhesive coating, heat sealing). The thus obtained films can be used in intended applications.

45 The laminated films of the present invention can be used in various applications by themselves, but may be laminated with a polyester film, a nylon film, a stretched polypropylene film or the like to improve the mechanical strengths, gas-barrier property, printability, etc.

The present invention is described in more detail below by way of Examples. However, the present invention is by no means restricted by the Examples.

50 In the following Examples of the present invention, individual test items were measured in accordance with the following methods.

[Tensile properties]

55 Tensile properties were measured in accordance with JIS Z 1702 and evaluated in the following four items, i.e. (1) tensile yield strength, Pa (Kgf/cm²), (2) tensile break strength, Pa (Kgf/cm²), (3) tensile elongation (%) and (4) tensile modulus, Pa (Kgf/cm²).

[Haze (%)]

Measured in accordance with ASTM D 1003.

[Gloss (%)]

Measured in accordance with ASTM D 2457.

[Elmendorf tear strength (kg/cm)]

Measured in accordance with JIS P 8116.

[Dart impact (g/66cm) [g/26 in]]

Measured in accordance with ASTM D 1709.

[Heat-sealing temperature]

A sample was subjected to heat sealing at various temperatures. A lowest heat-sealing temperature at which an adhesion strength capable of giving rise to material breakage was obtained, was taken as a heat-sealing temperature of the sample.

[Wettability]

A film which had been subjected to a corona treatment at a voltage of 100 V, was coated with various wetting index standard solutions (manufactured by Kishida Kagaku K.K.) each having a numerical symbol, at a width of 1 cm. After 2 seconds, those standard solutions remaining at the original width (1 cm) were examined and the largest numerical symbol of these solutions was taken as the wettability (10^{-3} N) of the film. (In a film having a small wettability, the width of coated solution became narrower than 1 cm after 2 seconds owing to the interfacial tension between the film and the coated solution.)

[Adhesivity]

A commercial cellophane tape (manufactured by Nichiban K.K.) was adhered to the corona-treated (100 V) surface of a film to prepare a sample for adhesivity. The sample was tested for adhesion strength by T-peel (crosshead speed: 200 mm/min, sample width: 24 mm).

Examples 1-4

[Preparation of resin composition for layer A]

As the resin composition constituting the layer A, there was used a resin composition prepared by mixing, at a 50/50 weight ratio, an amorphous polyolefin [REXTAC RT2780 (trade name) manufactured by Rexene Co.] having a density of 0.86 g/cm³, a propylene content of 65% by weight, a butene-1 content of 35% by weight and a melt viscosity of 10 Pa.s (10,000 cp) (190° C) and a crystalline polypropylene [B301H (trade name) manufactured by Ube Industries, Ltd.] having a density of 0.90 g/cm³ and a melt flow rate (MFR) (230° C) of 1.0 g/10 min and then melt kneading the mixture at 200 °C for 30 minutes.

[Molding of laminated films]

There were used, for the layer A, the resin composition prepared above and, for the B₁ layer, a crystalline polypropylene [RF395 (trade name) manufactured by Ube Industries, Ltd.] having a density of 0.91 g/cm³, a melt flow rate (MFR) (230° C) of 9 g/10 min and a melting point of 138° C. The resin composition and the crystalline polypropylene were subjected to melt three-layer co-extrusion using three independent extruders and a three-layer die connected to the extruders, so as to give a B₁/A/B₁ thickness ratio shown in Table 1; the resulting laminate was subjected to airing and quenching with a water-cooled sizing ring; the cooled laminate was subjected to blow-up molding into a tube-shaped film; the film was immediately wound, whereby three-layered laminated films each having a total thickness shown in Table 1 were obtained. The three-layered laminated films were measured for properties. The results are shown in Table 1.

Film Structure			Film Properties										Haze (%)	Gloss (%)	Dart Impact g/66cm (g/26 inch)
Total Thick- ness (μ)	Ratio of Layer Thick- ness B ₁ /A/B ₂	Tensile Yield Strength 10 ³ Pa (Kg/cm ²)		Tensile Break Strength 10 ³ Pa (Kg/cm ²)		Tensile Elongation (23°C) (%)		Tensile Modulus 10 ³ Pa (Kg/cm ²)		Elmendorf Tear Strength (Kg/cm)					
		*1	*2	*1	*2	*1	*2	*1	*2	*1	*2				
1	50	1/4/1	101 (103)	97 (99)	390 (398)	333 (340)	786 (851)	1603 (1635)	1810 (1846)	79	210	0.5	131	878	
2	50	1/2/1	117 (119)	108 (110)	447 (456)	347 (354)	769 (815)	1792 (1827)	1814 (1850)	56	217	0.5	135	878	
3	100	1/4/1	103 (105)	100 (102)	360 (367)	322 (328)	811 (828)	2303 (2348)	2206 (2249)	102	128	0.5	161	1229	
4	100	1/2/1	119 (121)	115 (117)	402 (410)	366 (373)	816 (823)	2299 (2344)	2256 (2300)	86	119	0.5	165	1141	
5	50	1/4/1	88 (90)	86 (88)	346 (353)	304 (310)	810 (840)	1520 (1550)	1643 (1675)	85	231	0.4	132	850	
6	50	1/4/1	118 (120)	113 (115)	428 (436)	368 (375)	815 (853)	2501 (2550)	2599 (2650)	53	92	0.7	136	658	
7	100	1/2/2	135 (138)	127 (129)	464 (473)	398 (406)	798 (853)	2599 (2650)	2854 (2910)	90	228	2.0	131	≥1320	
8	250	1/5/1	78 (80)	78 (80)	255 (260)	255 (260)	900	1373 (1400)	1373 (1400)	≥107	≥107	1.5	152	≥1320	
9	60	1/2/1	121 (123)	119 (121)	461 (470)	375 (382)	790	1618 (1650)	1648 (1680)	124	160	0.7	145	NB	
10	60	1/2/1	112 (114)	100 (102)	453 (462)	354 (361)	780	1471 (1500)	1373 (1400)	128	156	1.0	135	NB	
11	60	1/2/1	135 (138)	133 (136)	473 (482)	391 (399)	713	1687 (1720)	1755 (1790)	138	169	2.5	138	NB	

Table 1

Film Structure			Film Properties											Dart Impact g/66cm (9/26 inch)		
Total Thickness (μ)	Ratio of Layer Thickness $B_1/A/B_2$	Tensile Yield Strength (23°C) 10 ⁵ Pa (Kgf/cm ²)		Tensile Break Strength (23°C) 10 ⁵ Pa (Kgf/cm ²)		Tensile Elongation (23°C) (%)		Tensile Modulus (23°C) 10 ⁵ Pa (Kgf/cm ²)		Elmendorf Tear Strength (Kg/cm)	Haze (%)	Gloss (%)				
		*1	*2	*1	*2	*1	*2	*1	*2				*1		*2	
Comparative Examples	1	50	-	110 (112)	106 (108)	239 (244)	248 (253)	384	645	1453 (1482)	1594 (1625)	52	65	6.8	88	172
	2	50	-	136 (139)	128 (131)	414 (422)	399 (407)	779	1027	1899 (1936)	2125 (2167)	30	168	7.7	104	142
	3	50	-	83 (85)	66 (67)	408 (416)	389 (397)	400	980	794 (810)	803 (819)	187	290	7.0	30	315
	4	60	-	51 (52)	50 (51)	309 (315)	300 (306)	515	680	520 (530)	584 (595)	42	52	2.2	137	562
	5	50	-	298 (304)	49 (50)	582 (593)	309 (315)	487	4	4072 (4152)	4180 (4262)	5	480	72.9	7	20
	6	60	-	155 (158)	145 (148)	499 (509)	466 (475)	807	872	3393 (3460)	3556 (3626)	15	24	0.8	150	369

Table 1 (continued)

Example 5

A three-layered laminated film having a total thickness shown in Table 1 was obtained in the same manner as in

Example 1 except that the mixing ratio of the amorphous polyolefin and the crystalline polypropylene was 70/30 in the resin composition for the layer A. The three-layered laminated film was measured for properties. The results are shown in Table 1.

5 Example 6

A three-layered laminated film having a total thickness shown in Table 1 was obtained in the same manner as in Example 1 except that the mixing ratio of the amorphous polyolefin and the crystalline polypropylene was 30/70 in the resin composition for the layer A. The three-layered laminated film was measured for properties. The results are shown in Table 1.

Examples 7-8

[Resin composition for layer A]

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As the resin constituting the layer A, there was used an amorphous polypropylene (boiling-n-heptane insoluble = 46%, number-average molecular weight = 48,000) having a density of 0.88 g/cm³ and an MFR (230°C) of 8.4 g/10 min.

[Constitution of laminated films]

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Example 7: There were used, for the layer A, the above resin composition and, for the layer B₁, a crystalline polypropylene [FL453 (trade name) manufactured by Ube Industries, Ltd.] having a density of 0.90 g/cm³, an MFR (230°C) of 2.7 g/10 min and a melting point of 139°C. The layer constitution was B₁/A/B and the ratio of layer thicknesses was as shown in Table 1.

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Example 8: There were used, for the layer A, the above resin composition and, for the layer B₁, a crystalline polypropylene [RF395 (trade name) manufactured by Ube Industries, Ltd.] having a density of 0.91 g/cm³, an MFR (230°C) of 9.0 g/10 min and a melting point of 138°C. The layer constitution was B₁/A/B₁ and the ratio of layer thicknesses was as shown in Table 1.

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[Molding of laminated films]

The resin composition and the crystalline polypropylene were subjected to melt three-layer co-extrusion using three independent extruders and a three-layer die connected to the extruders, so as to give the above film constitution; the resulting laminate was subjected to airing and quenching with a water-cooled sizing ring; the cooled laminate was subjected to blow-up molding into a tube-shaped film; the film was immediately wound, whereby three-layered laminated films each having a total thickness shown in Table 1 were obtained.

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The three-layered laminated films were measured for properties. The results are shown in Table 1.

Comparative Example 1

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As the material for film, there was used a low-density polyethylene (LDPE) [F019 (trade name) manufactured by Ube Industries, Ltd.] having a density of 0.92 g/cm³ and a melt index (MI) (190°C) of 0.9 g/10 min. The material was passed through an extruder and an inflation film molding apparatus with a ring-shaped die to wind a tube-shaped film to obtain a LDPE single-layered film of 50 μm in thickness. The film was measured for properties. The results are shown in Table 1.

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Comparative Examples 2-5

Using, as the film materials, a linear low-density polyethylene (LLDPE) [FB123M (trade name) manufactured by Ube Industries, Ltd.] having a density of 0.92 g/cm³ and an MI (190°C) of 0.8 g/10 min in Comparative Example 2, a very-low-density polyethylene (VLDPE) [Z517A (trade name) manufactured by Ube Industries, Ltd.] having a density of 0.91 g/cm³ and an MI (190°C) of 1.5 g/10 min in Comparative Example 3, an ethylene/vinyl acetate copolymer (EVA) [V215 (trade name) manufactured by Ube Industries, Ltd.] having a density of 0.93 g/cm³, an MI (190°C) of 2.0 g/10 min and a vinyl acetate content of 15% by weight in Comparative Example 4, and a high-density polyethylene (HDPE) [6000 (trade name) manufactured by Tosoh Corporation] having a density of 0.96 g/cm³ and an MI (190°C) of 0.1 g/10 min in Comparative Example 5, there were obtained a LLDPE single-layered film (Comparative Example 2), a VLDPE single-layered film (Comparative Example 3), an EVA single-layered film (Comparative Example 4) and a HDPE single-layered film (Comparative Example 5) each having a thickness shown in Table 1, in the same manner as in Comparative Example 1. Each film was measured for properties. The results are shown in Table 1.

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Comparative Example 6

A film of 60 μm in thickness was obtained in the same manner as in Example 1 except that the same crystalline polypropylene as used for the layer B₁ in Example 1 was subjected to three-layer co-extrusion using the same molding apparatus for three-layered laminated film, as used in Example 1. The film was measured for properties. The results are shown in Table 1.

Example 9

A three-layered laminated film having a total thickness shown in Table 1 was obtained in the same manner as in Example 1. The three-layered laminated film was measured for properties. The results are shown in Table 1.

Example 10

A three-layered laminated film having a total thickness shown in Table 1 was obtained in the same manner as in Example 1 except that in the resin used in the layer A, the mixing ratio of the amorphous polypropylene and the crystalline polypropylene was changed to 7/3. The three-layered laminated film was measured for properties. The results are shown in Table 1.

Example 11

A three-layered laminated film having a total thickness shown in Table 1 was obtained in the same manner as in Example 1 except that the resin composition for layer A was prepared by mixing the same crystalline polypropylene (B301H) as used in Example 1 and a polybutene [M2481 (trade name) manufactured by Mitsui Petrochemical Industries, Ltd.] at a ratio of 50/50 and then melt-kneading the mixture using an extruder and that said resin composition was used as an intermediate layer of the laminated film. The three-layered laminated film was measured for properties. The results are shown in Table 1.

As is clear from Examples 1-11 and Comparative Examples 1-6, the three-layered laminated films of the present invention each consisting of one layer A and two layer B₁, as compared with the films of Comparative Examples 1-6, are well balanced in tensile elongation and other tensile properties, have flexibility and excellent mechanical strength, and have good transparency, haze and gloss in view of the haze and gloss values.

Example 12

[Preparation of resin composition for layer A]

A resin composition was obtained in the same manner as in Example 1.

[Molding of laminated film]

There were used, for the layer A, the resin composition prepared above and, for the layer B₂, a low density polyethylene [F200 (trade name) manufactured by Ube Industries, Ltd.] having a density of 0.92 g/cm³, a Melt Index (MI) (190°C) of 2 g/10 min.

The resin composition and the crystalline polypropylene were subjected to melt three-layer co-extrusion using three independent extruders and a three-layer die connected to the extruders, so as to give a B₂/A/B₂ thickness ratio shown in Table 2; the resulting laminate was subjected to airing and quenching with a watercooled sizing ring; the cooled laminate was subjected to blow-down molding into a tube-shaped film; the film was immediately wound, whereby a three-layered laminated film having a total thickness shown in Table 2 were obtained. The three-layered laminated film was measured for properties. The results are shown in Table 3. The composition of the film is shown in Table 2.

Example 13

A three-layered laminated film having a total thickness shown in Table 2 was obtained in the same manner as in Example 12 except that as the layer B₂ there was used a linear low-density polyethylene (LLDPE) [FA120N (trade name) manufactured by Ube Industries, Ltd.] having a density of 0.92 g/cm³ and an MI (190°C) of 1.9 g/10 min. The three-layered laminated film was measured for properties. The results are shown in Table 3. The composition of the film is shown in Table 2.

Example 14

A three-layered laminated film having a total thickness shown in Table 2 was obtained in the same manner as in Example 12 except that as the layer B₂ there was used a very-low-density polyethylene (VLDPE) [Z522 (trade name) manufactured by Ube Industries, Ltd.] having a density of 0.895 g/cm³ and an MI (190°C) of 3.0 g/10 min. The three-layered laminated film was measured for properties. The results are shown in Table 3. The composition of the film is shown in Table 2.

Example 15

A three-layered laminated film having a total thickness shown in Table 2 was obtained in the same manner as in Example 12 except that as the layer B₂ there was used an ethylene/vinyl acetate copolymer (EVA) [V215 (trade name) manufactured by Ube Industries, Ltd.] having a density of 0.93 g/cm³, an MI (190°C) of 2.0 g/10 min and a vinyl acetate content of 15% by weight. The three-layered laminated film was measured for properties. The results are shown in Table 3. The composition of the film is shown in Table 2.

Comparative Example 7

As the material for film, there was used an ethylene/vinyl acetate copolymer (EVA) [V215 (trade name) manufactured by Ube Industries, Ltd.] having a density of 0.93 g/cm³, an MI (190°C) of 2.0 g/10 min and a vinyl acetate content of 15% by weight. The material was passed through an extruder and an inflation film molding apparatus with a ring-shaped die to wind a tube-shaped film to obtain an EVA single-layered film of 60 µm in thickness. The film was measured for properties. The results are shown in Table 3.

Comparative Example 8

A LDPE single-layered film was obtained in the same manner as in Comparative Example 7 except that as the material for film there was used a low-density polyethylene [F200 (trade name) manufactured by Ube Industries, Ltd.] having a density of 0.92 g/cm³ and an MI (190°C) of 2 g/10 min. The film was measured for properties. The results are shown in Table 3.

Comparative Example 9

A LLDPE single-layered film was obtained in the same manner as in Comparative Example 7 except that as the material for film there was used a linear low-density polyethylene [FA120N (trade name) manufactured by Ube Industries, Ltd.] having a density of 0.92 g/cm³ and an MI (190°C) of 1.0 g/10 min. The film was measured for properties. The results are shown in Table 3.

Comparative Example 10

A PP single-layered film was obtained in the same manner as in Comparative Example 7 except that as the material for film there was used a crystalline polypropylene (PP) [RF395 (trade name) manufactured by Ube Industries, Ltd.] having a density of 0.91 g/cm³, an MFR (230 °C) of 9 g/10 min and a melting point of 138 °C. The film was measured for properties. The results are shown in Table 3.

Table 2

		Film Structure		Film Composition	
		Total Thickness (μ)	Ratio of Layer Thickness (B_2)/(A)/(B ₂)	A Layer	B ₂ Layer
Examples	12	60	1 / 2 / 1	Amorphous polyolefin/PP =50/50	LDPE
	13	60	1 / 2 / 1	Amorphous polyolefin/PP =50/50	LLDPE
	14	60	1 / 2 / 1	Amorphous polyolefin/PP =50/50	VLDPE
	15	60	1 / 2 / 1	Amorphous polyolefin/PP =50/50	EVA
Comparative Examples	7	60	—	EVA	
	8	60	—	LDPE	
	9	60	—	LLDPE	
	10	60	—	PP	

Note : PP refers to a polypropylene
 LDPE refers to a low-density polyethylene
 LLDPE refers to a linear low-density polyethylene
 VLDPE refers to a very-low-density polyethylene
 EVA refers to an ethylene/vinyl acetate copolymer

Table 3

	Film Properties									
	Tensile Strength (23°C)		Tensile Break Strength (23°C)		Tensile Elongation (23°C)		Tensile Modulus (23°C)		Tensile Modulus (80°C)	
	•1	•2	•1	•2	•1	•2	•1	•2	•1	•2
Examples	12	—	740 (245)	220 (224)	610	840	994 (1014)	1028 (1048)	298 (304)	327 (328)
	13	76 (77)	919 (423)	298 (304)	870	1040	951 (972)	1051 (1072)	298 (292)	284 (284)
	14	54 (55)	293 (299)	28 (299)	820	820	529 (539)	533 (544)	119 (121)	132 (135)
	15	70 (71)	231 (236)	28 (232)	690	690	111 (113)	114 (116)	14 (14)	15 (15)
	7	51 (52)	309 (315)	300 (316)	515	680	520 (530)	581 (595)	281 (281)	281 (281)
Comparative Examples	8	—	21 (225)	22 (226)	360	620	1615 (1677)	1791 (1826)	386 (392)	393 (401)
	9	131 (134)	257 (262)	28 (253)	570	730	2157 (2200)	2530 (2580)	581 (611)	599 (611)
	10	155 (158)	499 (509)	466 (475)	807	872	3393 (3460)	3536 (3626)	817 (833)	817 (833)

•Note* (1) NB refers to "not broken"
 (2) Measurement was impossible because the sample was in a molten state.

*1 denotes the lengthwise direction
 *2 denotes the crosswise direction

As is clear from Examples 12-15 and Comparative Examples 7-10, the three-layered laminated films each consisting of one layer A and two layers B₂, as compared with the films of Comparative Examples 7-10, are well balanced in flexibility, heat resistance and mechanical strengths and are sealable at low temperatures.

Example 16

[Resin composition for layer A]

As the resin composition constituting the layer A, there was used the resin composition prepared in Example 1.

[Molding of laminated film]

There were used, for the layer A, the resin composition prepared above and, for the layer B₃, a resin composition obtained by mixing a crystalline polypropylene [RF395 (trade name) manufactured by Ube Industries, Ltd.] having a density of 0.91 g/cm³, an MFR (230°C) of 9 g/10 min and a melting point of 138°C and an ethylene/vinyl acetate copolymer (EVA) [Z289 (trade name) manufactured by Ube Industries, Ltd.] having an MI (190°C) of 150 and an vinyl acetate content of 28%, at a weight ratio of 90/10 and then melt-kneading the mixture using an extruder.

The two resin compositions were subjected to melt three-layer co-extrusion using three independent extruders and a three-layer die connected to the extruders, so as to give a B₃/A/B₃ thickness ratio shown in Table 4; the resulting laminate was subjected to airing and quenching with a water-cooled sizing ring; the cooled laminate was subjected to blow-down molding into a tube-shaped film; the film was immediately wound, whereby a three-layered laminated film having a total thickness shown in Table 4 was obtained. The three-layered laminated film was measured for properties. The results are shown in Table 4. The composition of the film is shown in Table 5.

Example 17

A three-layered laminated film having a total thickness shown in Table 4 was obtained in the same manner as in Example 16 except that in the resin composition for the layer B₃, the mixing ratio of the crystalline polypropylene and the ethylene/vinyl acetate copolymer was changed to 80/20. The three-layered laminated film was measured for properties. The results are shown in Table 4. The composition of the film is shown in Table 5.

Example 18

A three-layered laminated film having a total thickness shown in Table 4 was obtained in the same manner as in Example 16 except that in the resin composition for the layer B₃, the mixing ratio of the amorphous polypropylene and the crystalline polypropylene was changed to 70/30. The three-layered laminated film was measured for properties. The results are shown in Table 4. The composition of the film is shown in Table 5.

Example 19

A three-layered laminated film having a total thickness shown in Table 4 was obtained in the same manner as in Example 18 except that in the resin composition for the layer B₃, the mixing ratio of the crystalline polypropylene and the ethylene/vinyl acetate copolymer was changed to 85/15. The three-layered laminated film was measured for properties. The results are shown in Table 4. The composition of the film is shown in Table 5.

Example 20

A three-layered laminated film having a total thickness shown in Table 4 was obtained in the same manner as in Example 18 except that in the resin composition for the layer B₃, the mixing ratio of the crystalline polypropylene and the ethylene/vinyl acetate copolymer was changed to 80/20. The three-layered laminated film was measured for properties. The results are shown in Table 4. The composition of the film is shown in Table 5.

Example 21

A three-layered laminated film having a total thickness shown in Table 4 was obtained in the same manner as in Example 13 except that the resin composition for layer A was prepared by mixing the same crystalline polypropylene as used in Example 16 and a polybutene [M2481 (trade name) manufactured by Mitsui Petrochemical Industries, Ltd.] at a ratio of 50/50 and then melt-kneading the mixture using an extruder and that said resin composition was used as an intermediate layer of the laminated film. The three-layered laminated film was measured for properties. The results are shown in Table 4. The composition of the film is shown in Table 5.

Table 4

Film Structure			Film Properties													
Comp Ex	11	60	—	Total thickness (μ)	Ratio of Layer Thickness (B ₃)/(A)/(B ₃)	Tensile Yield Strength (23°C) 10 ⁵ (kg/cm ²)		Tensile Break Strength (23°C) 10 ⁵ (kg/cm ²)		Tensile Elongation (23°C) (%)	Tensile Modulus (23°C) 10 ⁵ Po(kg/cm ²)	Wettability (10 ⁻³ N)	Adhesivity (g/24 mm)	Haze (%)		
						*1	*2	*1	*2							
Examples	16	60	1 / 2 / 1			120 (0.22)	105 (0.07)	411 (419)	314 (320)	777	867	1520 (1550)	1441 (1520)	43	580	0.7
	17	60	1 / 2 / 1			100 (0.02)	102 (0.02)	391 (393)	287 (293)	1037	893	1255 (1280)	1005 (1025)	45	650	5.2
	18	60	1 / 2 / 1			115 (0.17)	101 (0.03)	440 (449)	333 (340)	871	910	1285 (1320)	1035 (1300)	43	570	1.3
	19	60	1 / 2 / 1			116 (0.18)	101 (0.03)	424 (415)	285 (291)	903	843	1173 (1200)	1108 (1130)	45	610	1.7
	20	60	1 / 2 / 1			101 (0.03)	103 (0.04)	394 (395)	250 (255)	798	791	1157 (1180)	1064 (1110)	46	660	4.3
	21	60	1 / 2 / 1			129 (0.31)	124 (0.26)	413 (431)	320 (330)	716	802	1627 (1700)	1518 (1630)	43	570	2.8
	11	60	—			155 (0.50)	125 (0.40)	474 (500)	356 (473)	807	872	3313 (3460)	3056 (3620)	40	480	0.8

*1 denotes the lengthwise direction

*2 denotes the crosswise direction

Table 5

		Film Composition	
		A Layer	B ₃ Layer
Examples	16	B301H/RT2780 =50/50	RF395/Z289 =90/10
	17	B301H/RT2780 =50/50	RF395/Z289 =80/20
	18	B301H/RT2780 =30/70	RF395/Z289 =90/10
	19	B301H/RT2780 =30/70	RF395/Z289 =85/15
	20	B301H/RT2780 =30/70	RF395/Z289 =80/20
	21	B301H/M2481 =50/50	RF395/Z289 =90/10

As is clear from Examples 16-21, the three-layered laminated films of the present invention each consisting of one layer A and two layers B₃, are flexible and have excellent wettability and adhesivity.

As described above, the laminated films of the present invention are soft films which have good transparency, haze and gloss, which have excellent flexibility and mechanical strengths, and which have high impact resistance, surface wettability and low-temperature sealability. Therefore, the present laminated films are applicable to various usages in place of soft vinyl chloride films which are raising social problems such as environmental pollution.

Claims

1. A laminated film obtainable by melt coextrusion of at least 2 layers comprising:
 - 5 a layer (A) containing a fibre-free composition comprising (i) 30 to 100% by weight of an amorphous polyolefin derived from, by weight of the polyolefin, at least 50% of propylene and/or butene-1, and (ii) 70 to 0% by weight of a crystalline polypropylene; and
 - an outermost layer (B) comprising (i) a crystalline polypropylene, (ii) an ethylene (co)polymer or (iii) a blend of a crystalline polypropylene and a copolymer of ethylene and a vinyl monomer having a polar group, or a polymer that is obtainable by grafting a vinyl monomer having a polar group to an ethylene (co)polymer.
2. A film according to claim 1, wherein the vinyl monomer is vinyl acetate, ethyl acrylate, methyl acrylate or maleic anhydride.
- 15 3. A film according to claim 1 or claim 2, wherein layer B contains said blend of the crystalline polypropylene and the ethylene copolymer or graft polymer, in a weight ratio of from 98:2 to 50:50.
4. A film according to claim 1, wherein layer B contains an ethylene (co)polymer having a Shore D hardness (23°C) of 50 or less.
- 20 5. A film according to any preceding claim, whose two outermost layers are each a layer (B).
6. A film according to any preceding claim, wherein the amorphous polyolefin has a boiling-n-heptane insoluble content of 70% by weight or less and a number-average molecular weight of 1,000 to 200,000.
- 25 7. A film according to any preceding claim, wherein the amorphous polyolefin is derived from propylene as the main component, and is polypropylene or a propylene/ethylene, propylene/butene-1, propylene/hexene-1, propylene/butene-1/ethylene, propylene/hexene-1/octene-1 or propylene/hexene-1/4-methylpentene-1 copolymer.
- 30 8. A film according to any of claims 1 to 6, wherein the amorphous polyolefin is derived from butene-1 as the main component, and is polybutene-1 or a butene-1/ethylene, butene-1/propylene, butene-1/propylene/ethylene, butene-1/hexene-1/octene-1 or butene-1/hexene-1/4-methylpentene-1 copolymer.
- 35 9. A film according to any preceding claim, wherein layer A comprises a crystalline polypropylene selected from propylene homopolymer, a propylene/ethylene random or block copolymer containing the ethylene component in an amount of 1 to 30% by weight, and a propylene/butene-1 random or block copolymer containing the butene-1 component in an amount of 1 to 20% by weight.
- 40 10. A film according to any preceding claim, which is monoaxially or biaxially stretched.

Patentansprüche

1. Laminatfolie, erhältlich durch Schmelz-Coextrusion von mindestens 2 Schichten umfassend:
 - 45 eine Schicht (A), die eine faserfreie Zusammensetzung enthält, welche (i) 30 bis 100 Gew.-% eines amorphen Polyolefins, abgeleitet von, bezogen auf das Gewicht des Polyolefins, mindestens 50% Propylen und/oder Buten-1, und (ii) 70 bis 0 Gew.-% eines kristallinen Polypropylen umfasst; und
 - eine äußerste Schicht (B), welche (i) ein kristallines Polypropylen, (ii) ein Ethylen-(Co)polymer oder (iii) eine Mischung eines kristallinen Polypropylen und eines Copolymers von Ethylen und einem Vinylmonomer mit einer polaren Gruppe oder eines Polymers, das durch Pfropfen eines Vinylmonomers mit einer polaren Gruppe auf ein Ethylen-(Co)polymer erhalten werden kann, umfasst.
- 50 2. Folie nach Anspruch 1, worin das Vinylmonomer Vinylacetat, Ethylacrylat, Methylacrylat oder Maleinsäureanhydrid ist.
- 55 3. Folie nach Anspruch 1 oder Anspruch 2, worin Schicht B die Mischung des kristallinen Polypropylen und des Ethylen-Copolymers oder -Pfropfpolymer in einem Gewichtsverhältnis von 98:2 bis 50:50 enthält.
4. Folie nach Anspruch 1, worin Schicht B ein Ethylen-(Co)polymer mit einer Shore D-Härte (23°C) von 50 oder weni-

ger enthält.

5. Folie nach irgendeinem vorhergehenden Anspruch, deren zwei äußerste Schichten jeweils eine Schicht (B) sind.
- 5 6. Folie nach irgendeinem vorhergehenden Anspruch, worin das amorphe Polyolefin einen Gehalt an in siedendem n-Heptan unlöslichem Material von 70 Gew.-% oder weniger und ein Zahlenmittel des Molekulargewichts von 1000 bis 200000 aufweist.
- 10 7. Folie nach irgendeinem vorhergehenden Anspruch, worin das amorphe Polyolefin von Propylen als Hauptkomponente abgeleitet ist und Polypropylen oder ein Propylen/Ethylen-, Propylen/Buten-1-, Propylen/Hexen-1-, Propylen/Buten-1/Ethylen-, Propylen/Hexen-1/Octen-1- oder Propylen/Hexen-1/4-Methylpenten-1-Copolymer ist.
- 15 8. Folie nach irgendeinem der Ansprüche 1 bis 6, worin das amorphe Polyolefin von Buten-1 als Hauptkomponente abgeleitet ist und Polybuten-1 oder ein Buten-1/Ethylen-, Buten-1/Propylen-, Buten-1/Propylen/Ethylen-, Buten-1/Hexen-1/Octen-1- oder Buten-1/Hexen-1/4-Methylpenten-1-Copolymer ist.
- 20 9. Folie nach irgendeinem vorhergehenden Anspruch, worin Schicht A ein aus Propylen-Homopolymer, einem statistischen Copolymer oder Blockcopolymer von Propylen/Ethylen, das die Ethylen-Komponente in einer Menge von 1 bis 30 Gew.-% enthält, und einem statistischen Copolymer oder Blockcopolymer von Propylen/Buten-1, das die Buten-1-Komponente in einer Menge von 1 bis 20 Gew.-% enthält, ausgewähltes kristallines Polypropylen umfaßt.
10. Folie nach irgendeinem vorhergehenden Anspruch, welche monoaxial oder biaxial gereckt ist.

Revendications

- 25 1. Film stratifié pouvant être obtenu par coextrusion à l'état fondu d'au moins deux couches comprenant :
 - une couche (A) contenant une composition exempte de fibres comprenant (i) 30 à 100 % en poids d'une polyoléfine amorphe obtenue à partir d'au moins 50 % de propylène et/ou de butène-1, en poids de la polyoléfine,
 - 30 et (ii) de 70 à 0 % en poids d'un polypropylène cristallin ; et
 - une couche la plus externe (B) comprenant (i) un polypropylène cristallin, (ii) un (co)polymère de l'éthylène ou (iii) un mélange d'un polypropylène cristallin et d'un copolymère de l'éthylène et d'un monomère vinylique ayant un groupe polaire, ou d'un polymère qui peut être obtenu en greffant un monomère vinylique ayant un groupe polaire sur un (co)polymère de l'éthylène.
- 35 2. Film selon la revendication 1, dans lequel le monomère vinylique est l'acétate de vinyle, l'acrylate d'éthyle, l'acrylate de méthyle ou l'anhydride maléique.
- 40 3. Film selon les revendications 1 ou 2, dans lequel la couche (B) contient ce mélange du polypropylène cristallin et du copolymère de l'éthylène ou du polymère greffé, dans un rapport pondéral de 98:2 à 50:50.
4. Film selon la revendication 1, dans lequel la couche (B) contient un (co)polymère de l'éthylène ayant une dureté Shore D (à 23 °C) de 50 ou moins.
- 45 5. Film selon l'une quelconque des revendications précédentes, dont les deux couches les plus externes sont chacune une couche (B).
- 50 6. Film selon l'une quelconque des revendications précédentes, dans lequel la polyoléfine amorphe a une teneur en matières insolubles dans le n-heptane bouillant de 70 % en poids ou moins et une masse moléculaire moyenne en nombre de 1 000 à 200 000.
- 55 7. Film selon l'une quelconque des revendications précédentes, dans lequel la polyoléfine amorphe est obtenue à partir de propylène comme constituant principal, et est le polypropylène ou un copolymère propylène/éthylène, propylène/butène-1, propylène/hexène-1, propylène/butène-1/éthylène, propylène/hexène-1/octène-1 ou propylène/hexène-1/4-méthylpentène-1.
8. Film selon l'une quelconque des revendications 1 à 6, dans lequel la polyoléfine amorphe est obtenue à partir de butène-1 comme constituant principal et est un polybutène-1 ou un copolymère butène-1/éthylène, butène-1/propylène, butène-1/propylène/éthylène, butène-1/hexène-1/octène-1 ou butène-1/hexène-1/4-méthylpentène-1.

9. Film selon l'une quelconque des revendications précédentes, dans lequel la couche A comprend un polypropylène cristallin choisi parmi un homopolymère du propylène, un copolymère propylène/éthylène statistique ou séquencé contenant le constituant éthylène dans une proportion de 1 à 30 % en poids, et un copolymère propylène/butène-1 statistique ou séquencé contenant le constituant butène-1 dans une proportion de 1 à 20 % en poids.

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10. Film selon l'une quelconque des revendications précédentes, qui est étiré monoaxialement ou biaxialement.

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